Self-Sustaining Breeding in Advanced Reactors: Characterization of Selected Reactors

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Glossary

Ac Actinides, elements with atomic numbers 89 to 103.

AcCl Label for tetra-chloride salt of actinides $AcCl_4$.

Actinides Eigen-composition Stabilized actinides composition, an equilibrium composition which results from a long irradiation of one parent nuclide or their mixture. Necessary condition for the stabilization is that the mass of parent nuclide/s and the irradiation flux do not vary strongly. It is specific and inherent feature for every reactor and every parent nuclide/s and it represents the Eigen-vector of the respective Bateman equation.

B&B Breed-and-Burn; a mode of open cycle operation, where the fissile excess bred during irradiation is equal or higher than the discharged fissile mass after the irradiation. The reactor burns its own bred fuel. The discharged fuel does not need to be recycled and the fresh fuel does not need to contain primordial or synthetic fissile nuclides.

BG Breeding gain, BG = BR-1.

BR Breeding ratio, it is ratio between breeding rate and fission rate. In a self-sustaining breeder BR $\geq$ 1.

Breeding Transmutation of actinide nuclide initiated by neutron capture, which increases fission probability. Typically, primordial non-fissile (fertile) nuclides ^{232}Th and ^{238}U are transmuted into synthetic (man-made) fissile ^{233}U and ^{239}Pu.

Burnup Share of already fissioned actinides. It can be expressed in FIMA % (Fissions per Initial Metal Atom) or as energy released from given mass of actinides in MWd/kg (Megawatt days per kilogram). Since the energy per fission is approximately the same for all actinide nuclides, values expressed in these two units are proportional and roughly 10 times higher for the second unit.

Closed cycle A chain of process steps which is closed for the respective working medium. The medium, in nuclear fuel cycle the actinides, is recycled and does not extensively leave the chain.

Cross-section A measure of the probability that neutron will interact when flying by with the respective nucleus (microscopic cross-section) or when fling through with the respective nuclei concentration (macroscopic cross-section).

Equilibrium reactivity Reactivity provided by actinides Eigen-composition.

FPs Fission products, typically two fragments of actinides fission.

FLI Label for eutectic mixture of lithium fluoride salt and actinides tetra-fluoride salt LiF-AcF_4.

FLIBE Label for eutectic mixture of lithium fluoride and beryllium di-fluoride with addition of actinides tetra-fluoride salt LiF-BeF_2-AcF_4.

Irradiation chain Series of consecutive transmutations and of the respective daughter products induced by neutron irradiation of one parent nuclide, e.g., ^{232}Th or ^{238}U. The radioactive decays of the daughter products are part of the chain.
MA Minor actinides, synthetic actinides originated by uranium transmutation (Np, Am, Cm and higher elements) without Pu, which is some time called major actinide.
NaCl Label for eutectic mixture of sodium chloride salt and actinides tetra-chloride salt NaCl-$AcCl_4$.
Neutron economy Balance between neutron generation and neutron losses in a reactor. Good neutron economy has high neutron generation and small neutron losses by parasitic absorption and leakage from the reactor.
Open cycle A chain of process steps which is open for the respective working medium. The medium, in nuclear fuel cycle the actinides, is not recycled and leaves extensively the chain.
Primordial actinides Actinide nuclides, presumably originated by rapid neutron capture process during a supernova explosion, which have long enough half-life for radioactive decay to be still present on the earth.
Reactivity Reactivity is related to the effective (k_{eff}) or infinite (k_{inf}) neutron multiplication factor. Reactivity denotes the deviation from the self-sustaining fission chain reaction ($k = 1$), for which it is equal to zero. Reactor with negative reactivity ($k < 1$) cannot sustain the fission chain reaction. Positive reactivity ($k > 1$) means that there are more neutrons than needed for the self-sustaining fission chain reaction. From operational perspective, neutron absorber should compensate positive reactivity to control the chain reaction. From fuel cycle perspective, positive reactivity indicate that there are excess neutrons available for additional utilization by breeding or transmutations.
Self-sustaining breeding Breeding process where breeding rate $\geq$ fission rate, the mass of fissile synthetic nuclides is conserved in the reactor, and fission chain reaction is sustained solely by these synthetic nuclides. A long time-horizon is being considered in this article.
Subcritical/critical core A core that cannot/can sustain a fission chain-reaction.
Synthetic actinides Actinide nuclides originated, typically by neutron-induced transmutation, from primordial actinides. Synthetic nuclides are, by definition, a much broader group than minor actinides (MA) or trans-uranic elements. For thorium and uranium elements, both primordial and synthetic isotopes exist.
Waste Waste is unwanted side product, which is further unusable. In nuclear fuel cycle, synthetic actinides represent a side product, which is reusable. However, they can be declared as a waste by nuclear fuel cycle policy or when it is laborious to recover them, e.g., from reprocessing losses of certain fuel forms.

Abbreviations

FHR Fluoride high temperature reactor
GFR Gas cooled fast reactor
HPLWR High performance light water reactor
HTR High temperature reactor
LFR Lead cooled fast reactor
LWR Light water reactor
MCFR Molten chloride fast reactor
MFBR Metal fueled fast breeder reactor
MSFR Molten salt fast reactor
MSR Molten salt reactor (graphite moderated)
PHWR Pressurized heavy water reactor
RBMK Reaktor Bolshoi Moshnosti Kanalnyj (high-power graphite moderated water cooled channel-type reactor)
SCWR Supercritical Water-cooled Reactor
SFR Sodium cooled fast reactor

Introduction

This is the second of a series of three chapters that is dedicated to the identification of the advanced reactor technologies that are capable of self-sustaining breeding when fueled with either ^{232}Th or ^{238}U, and to the investigation of equilibrium fuel cycles properties and neutronics performance. The first chapter (Krepel, 2021a) characterizes the available fuel resources from the nuclear and reactor physics perspective. This second chapter determines the equilibrium fuel composition in advanced reactors and compares the basic features of the equilibrium fuel cycle and selected reactor neutronic performance characteristics at the equilibrium state. The third chapter (Krepel, 2021b) evaluates the fuel cycle performance from a neutronics perspective by means of the earlier

obtained equilibrium parameters. It provides an additional insight on why some advanced reactor concepts can act as self-sustaining breeders in closed or even open fuel cycle, while others do not.

Primordial actinide and self-sustaining breeding

The nuclear fuel cycle resources are not renewable and consist of three dominating primordial actinide nuclides: ^{235}U, ^{238}U and ^{232}Th. To fully utilize the potential of fertile ^{232}Th and ^{238}U, the self-sustaining breeding in closed cycle is needed. In such fuel cycle the primordial actinides are transmuted into synthetic fissile actinides ^{233}U and ^{239}Pu. This transmutation process is called breeding and reactors relying on this process are called breeders. Once originated, synthetic nuclides ^{239}Pu and ^{233}U can sustain the fission chain reaction and the breeding process itself. Accordingly, they practically act as a catalyzer for energy production from the primordial nuclides ^{238}U and ^{232}Th.

Simple fuel balance of the self-sustaining breeder requires that for each fissioned synthetic nucleus at least one transmutation of primordial nuclide takes place. Accordingly, up to two neutrons from each fission are needed; one for sustaining the chain reaction and one for maintaining of the fissile material balance. For each reactor and for each primordial nuclide, the composition of generated synthetic actinides tends to stabilize and reaches equilibrium concentration after sufficiently long neutron irradiation.

Irradiation chain and equilibrium cycle

Stabilized irradiation chain is obtained, when primordial nuclides ^{232}Th or ^{238}U are irradiated for sufficiently long time (Krepel, 2021a). The condition, necessary for this convergence, is that the primordial nuclide concentration and the irradiation flux do not vary. This condition is practically fulfilled by the operation of a breeder in closed fuel cycle, where all actinides are recycled and solely ^{232}Th or ^{238}U is refiled to the reactors as the feed material. The stabilized actinides composition is therefore also the Eigen-composition of the respective Bateman equation (Bateman, 1910). The so called equilibrium fuel cycle is a state, at which the self-sustaining breeder operated in closed cycle reaches the stabilized actinides composition. This equilibrium state is characterized by three variables: equilibrium fuel composition, equilibrium neutron spectrum and equilibrium multiplication factor.

The equilibrium fuel composition depends on the neutron spectrum and feed nuclide, here ^{232}Th or ^{238}U. The basic neutron spectrum type is determined by neutron scattering by the coolant and structural materials. However, it is also influenced by the equilibrium composition. Equilibrium spectrum and equilibrium composition together determine the equilibrium reactivity or actually equilibrium neutron multiplication factor (see **Fig. 1**). It is important to understand here that the multiplication factor does not affect the composition or spectrum. At the same time, reactivity excess or deficiency in equilibrium is the major indicator of the neutron economy and breeding capability.

From a historical perspective, the existence of an equilibrium state was revealed already during the pioneering time of nuclear energy, and the history is well described in Walter and Reynolds (1982). In a typical solid fuel fast breeder reactor, it takes five or six recyclings for plutonium vector stabilization (Journet et al., 1993). To enable concepts cross-comparison, a generalized Bateman equation was proposed and solved by a simplified approach in Salvatores et al. (2004). The term "equilibrium method" was also introduced in this reference. Inspired by this work, the EQL3D routine was developed (Krepel et al., 2008, 2012), which is a predecessor of the tool applied in the studies described in this article. The existence of an equilibrium state was obvious also for the developers of MSR concepts (MacPherson, 1985). Since it has liquid fuel, which enables continuous refueling and fission products removal, the knowledge of equilibrium was crucial for MSR. Several dedicated tools have been developed for MSR analyses in recent years, e.g., (Nuttin et al., 2005; Aufiero et al., 2013; Fiorina et al., 2013a; Seifried et al., 2013). The same is valid for bebble-bed HTR reactor, which also enables continuous removal and refueling of the fuel pebbles (Fratoni and Greenspan, 2007, 2010). Nowadays, there are many similar tools.

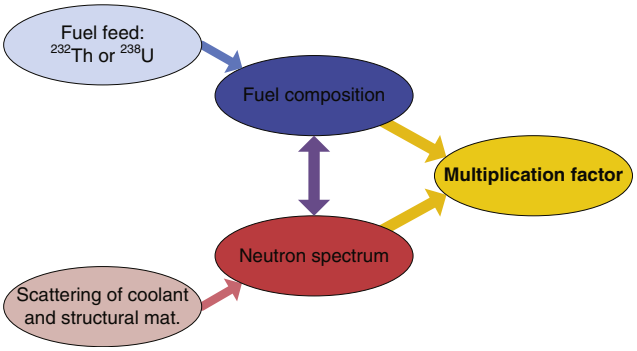

Fig. 1 Interdependency between neutron spectrum, fuel composition, and multiplication factor with indicated impact of the feed and scattering of coolant and structural materials. From Krepel J and Losa E (2019) Closed U-Pu and Th-U cycle in sixteen selected reactors: Comparison of major equilibrium features. *Annals of Nuclear Energy* 128: 341–357.

Objectives of this article

The objective of this article is to characterize the equilibrium fuel cycle for both the ^{232}Th and ^{238}U irradiation chains in 16 selected advanced reactors. The knowledge of the equilibrium state is of paramount importance for reactor performance evaluation, because it provides information:

- About the composition towards which the initial fuel tends to converge during irradiation;
- About the expected waste composition in both open and closed fuel cycles;
- Whether fuel parasitic neutron capture allows for establishing self-sustaining breeding;
- Whether there is sufficient neutron excess in the self-sustaining breeder for the utilization of legacy synthetic actinides (say, from LWR once-through fuel cycle) or for surplus fissile fuel breeding;
- About the rate of decrease of the legacy synthetic actinides concentration during irradiation.

This article is an extension and simplification of Krepel and Losa (2019), where details are provided. Selected aspect of this study have been published in Losa (2016), Krepel and Losa (2016, 2017) and Krepel et al. (2018).

Applied tool, assumptions and simulated reactors

The tool applied to more than 70 equilibrium calculations in this study is the Paul Scherrer Institut in-house procedure EQL0D (Hombourger et al., 2015, 2019). The simulations were relying on several simplifying assumptions, the major being Fission Products (FPs) neglection. The major impact of this simplification is an overestimation of the equilibrium reactivity. The selection of simulated reactors was aiming at covering all major neutron spectra and all major advanced reactor concepts. Accordingly, all Gen-IV reactor concepts described in this encyclopedia were included (Stanculescu, 2021).

Applied tool

The EQL0D procedure for burnup calculations is based on a MATLAB script. Historically there are three versions of the procedure: v1 is based on a MATLAB script and the deterministic ERANOS code (Rimpault, et al., 2002), and versions v2 and v3 that are based on a MATLAB script and the Monte Carlo code SERPENT 2 (Leppänen, 2013). The most advanced version v3 (Hombourger et al., 2016, 2019) adopts directly the burn-up matrix from the SERPENT code and applies the same method for the Bateman equation solution, including FPs. However, in this study the EQL0D v2 was applied (Hombourger, 2013; Losa, 2016), which relies on the MATLAB coupling with the SERPENT (see **Fig. 2**) code only through the actinide reaction rates. Hence, it cannot simulate FPs. As mentioned, the absence of FPs will result it equilibrium reactivity overestimation and slight spectrum softening. Whereas the spectral change can be neglected, the reactivity overestimation should be kept in mind.

The simulation of the ^{238}U and ^{232}Th irradiation used the ENDF/B-VII.0 nuclear data library (Chadwick, et al., 2006) and the SERPENT 2 code version was 2.1.23. The convergence of fuel composition was obtained after several iterations between the MATLAB burnup solver and the SERPENT code that generated spectrum-averaged cross-section for use in the burnup calculations. 20,000 source particles, 2000 active cycles, and 100 inactive cycles were used in the SERPENT calculations. The Monte Carlo nature of the code introduced issues with data noise. Its impact was not extensively evaluated for the EQL0D v2 version. Nevertheless, to minimize the oscillations relaxation between iterations was applied and relative mass change of ^{246}Cm smaller than 10^{-6} was used as a convergence criterion.

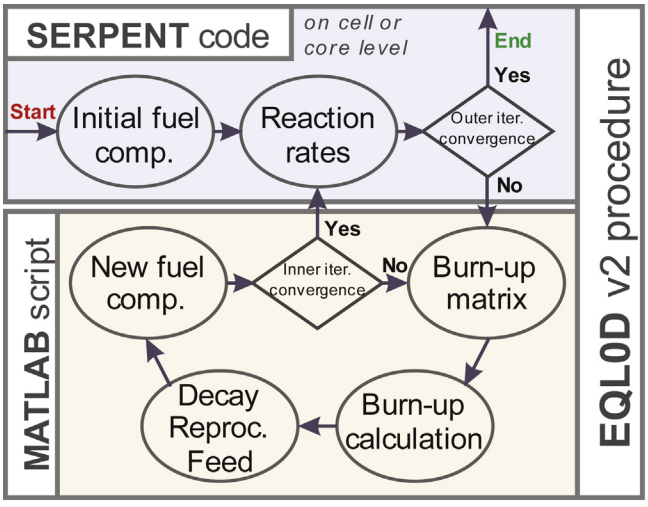

Fig. 2 Flow chart of EQL0D v2 routine. From Krepel J and Losa E (2019) Closed U-Pu and Th-U cycle in sixteen selected reactors: Comparison of major equilibrium features. *Annals of Nuclear Energy* 128: 341–357.

Assumptions

With the objective of this article being the comparison of the basic equilibrium features of the ^{232}Th and ^{238}U irradiation chains in advanced reactors, the results assembled here are based on three major assumptions:

(1) The amplitude of the neutron flux, in which the fuel is irradiated, is determined by the specific power. It is unimportant for the fuel composition convergence if the core is subcritical or supercritical.
(2) Each fission results in refilling of one respective ^{238}U or ^{232}Th atom, and FPs, as mentioned, are neglected. With this assumption, the usual composition variations during irradiation, cooling, and reprocessing stages are avoided.
(3) Neutron leakage is not accounted for. The core is simulated only on the assembly level with reflective boundary conditions.

The first assumption assures that equilibrium state is achieved independent from the multiplication factor. The second and third assumptions simplify the modeling; however, they also result in equilibrium reactivity over-prediction. Presence of FPs and of neutron leakage will definitely reduce the equilibrium reactivity. It will also have an impact on the reactor spectrum and thus on the equilibrium fuel composition. However, this impact is secondary and the fuel compositions obtained by these assumptions are sufficiently representative for the needs of this study.

Simulated reactors

The advanced reactor concepts evaluated in this study follow the Gen-IV systems SFR, VHTR, LFR, GFR, MSR, and SCWR introduced in this encyclopedia in Stanculescu (2021), Heidet (2021), Fütterer (2021), Alemberti (2021), Hatala (2021), Ignatiev (2021) and Huang and Zang (2021). However, this study includes also alternative concept such as the FHR (Fratoni, 2021), chlorides salt based MCFR (Krepel and Kramer, 2021), and advanced LWR (Shwageraus, 2021). Moreover, to ensure that the examined reactor concepts are providing maximal variability of the neutron spectrum, at least one example is adopted from concepts considered in this encyclopedia, specifically: pressurized water cooled reactors (Brown, 2021), heavy water cooled reactors (Nichita, 2021), gas cooled reactors, water cooled graphite moderated reactors, and liquid metal-cooled reactors. The boiling water cooled reactors (Hamon, 2021) is not included as such, but covered by a parametric moderation study for LWR. In general, three basic principles were used for the selection:

(1) All Gen-IV solid fuel reactors.
(2) Most realistic MSR concepts.
(3) Majority of existing power reactors.

As a result 16 reactor types were selected: 8 thermal and 8 fast; 6 based on liquid fuel in form of molten salts, and 10 featuring 4 different solid fuel types. The selected reactors and their labels used in results section are presented in **Table 1**. It also includes the coolant density steps for the LWR parametric study. The assumed specific powers in **Table 1** are determined by three fuel cycle parameters: average irradiation time, burnup and fuel density, and refer to the ratio of volumetric power and specific actinides density, with unit W/g_{Ac}. The specific power for the MSR cases, where the core size is not always known, was adopted from LWR for fluoride salts and from LFR for chlorides salts, respectively. The higher specific power proposed for MFBR is adopted from IGCAR (2004). The lattices of the selected reactors are illustrated in **Fig. 3**. Basically, there are five types of fuel: oxide pellets (LFR, SFR, LWR, RBMK, PHWR, HPLWR), TRISO coated particles in graphite pebbles in FHR and HTR, carbide pellets in GFR, zirconium based metallic fuel in MFBR, and molten salts in MSRs. The salts are further subdivided into chlorides and fluorides (see **Table 1**).

The solid fuel Gen-IV reactors include the sodium cooled reactor with both ceramic fuel (SFR) and metallic fuel (MFBR) options. The HTR is included together with its fluoride salt cooled version FHR. The other three reactors LFR, GFR, and SCWR, are included using one representative designs. Especially in case of SCWR, where HPLWR concept was selected, the choice is crucial, because the cladding material has strong impact on the core performance.

The MSR is rather a category of reactors than one single design; hence, the choice was driven by feasibility of the structural material, moderator, and fuel salt combinations. There are three major MSR classes: Graphite based MSRs, Homogeneous MSRs and Heterogeneous MSRs (IAEA, 2021). The latter class was excluded from this study. The reactors with heterogeneous layout rely on structural material to separate the fuel from dedicated coolant or non-graphite moderator. The major reason for their exclusion was the absence of publicly available data and some doubts about their technological maturity. However, a parametric study relevant to non-graphite moderated MSRs is presented in Hombourger (2018). The performance of heterogeneous fast MSRs is not addressed, as it is probable that the dedicated coolant and separation structural materials will increase both neutron scattering and capture reaction rates, and thus deteriorate the neutronics performance of this reactor type at the infinite lattice level considered in this study.

Only three MSR families were included in this lattice study: graphite moderated MSR, homogeneous fluoride fast MSFR, and homogenous chloride fast MCFR. Two fluoride and two chloride salts have been considered as the fuel carrier: ^{7}LiF-AcF_4 and ^{7}LiF-BeF_2-AcF_4 labelled as FLI and FLIBE and Na^{37}Cl-Ac$^{37}Cl_4$ and Ac$^{37}Cl_4$ labelled as NaCl and AcCl, respectively. The practical applicability of AcCl salt is rather hypothetical. The UCl_4 has reasonable melting temperature but is unstable; on the contrary, the stable UCl_3 has a high melting point. Since it is still much lower than the melting point of metallic actinides, AcCl was included in the study as a hypothetical actinide carrier delivering maximum performance. The selected salts are the best performing representatives from the neutronics perspective (Hombourger, 2018). For the graphite moderated MSR, the salt channel radius of 5 cm and the lattice geometry was adopted from Hombourger et al. (2016) using the case with 15% salt fraction (Rosenthal, et al., 1971; Krepel, et al., 2014).

Table 1 Parameters of 16 simulated reactors and cases for the parametric study (Krepel and Losa, 2019).

Reactor name	*Label*	*Fuel design adopted from (reference)*	*Fuel type*	*Specific power (W/g_{Ac})*
Solid fuel fast reactors				
European lead system	LFR	Consortium of EU FP7 LEADER project (Artioli et al., 2009; Krepel et al., 2012)	Oxide	54.8
European sodium fast reactor	SFR	Consortium of EU FP7 ESFR project (Buiron, et al., 2007; Krepel et al., 2012)	Oxide	48.8
Gas cooled fast reactor	GFR	Consortium of EU FP7 GoFastR project (Bosq et al., 2006; Perkó et al., 2015)	Carbide	40.1
Metal fueled fast breeder reactor	MFBR	IGCAR, Kalpakkam (IGCAR, 2004; Mohapatra, et al., 2013)	Metallic	178.6
Liquid fuel fast reactors				
Fast MSR: LiF-BeF_2-AcF_4 salt	MSFR-FLIBE	MSBR fuel salt properties (Rosenthal et al., 1971)	Fluoride salt	41.1
Fast MSR: LiF-AcF_4 salt	MSFR-FLI	Consortium of EU FP7 EVOL project (Heuer et al., 2014)	Fluoride salt	41.1
Fast MSR: NaCl-$AcCl_4$ salt	MCFR-NaCl	Salt eutectic comp. and density from Desyatnik et al. (1975) and Taube (1977)	Chloride salt	54.8
Fast MSR: $AcCl_4$ salt	MCFR-AcCl	Salt density adopted from Desyatnik et al. (1975) and Taube (1977)	Chloride salt	54.8
Solid fuel thermal reactors				
Fluoride high temperature reactor	FHR	University of California, Berkeley PB-AHTR (Fratoni et al., 2007)	Coated6particles	192.0
High temperature reactor	HTR	PBMR (Pty) Ltd. (IAEA, 2013)	Coated particles	96.8
Reaktor Bolshoi Moshnosti Kanalnyj	RBMK	NEA benchmark LEU-COMP-THERM-060 (OECD, 1995)	Oxide	13.7
Pressurized heavy water reactor	PHWR	CANDU benchmark (Pounders et al., 2011)	Oxide	32.1
High performance light water reactor	HPLWR	KIT Germany (Schulenberg, 2012)	Oxide	25.3
Light water reactor	LWR	VVER-1000 Tvel Russia (Emmett, 2000)	Oxide	41.1
Liquid fuel thermal reactors				
Thermal MSR: LiF-BeF_2-AcF_4 salt	MSR-FLIBE	Graphite share 85% (Hombourger et al., 2016) and MSFR salt (Rosenthal et al., 1971)	Fluoride salt	41.1
Thermal MSR: LiF-AcF_4 salt	MSR-FLI	Graphite share 85% (Hombourger et al., 2016) and MSFR salt (Heuer et al., 2014)	Fluoride salt	41.1
Parametric study				
Light water reactor	LWR	VVER-1000 Tvel Russia (Emmett, 2000)	Oxide	41.1
Cases: water density in % of nominal value	100, 95, 90, 85, 80, 75, 70, 65, 60, 55, 50, 45, 40, 35, 30, 25, 20, 15, 10, 5, 0			

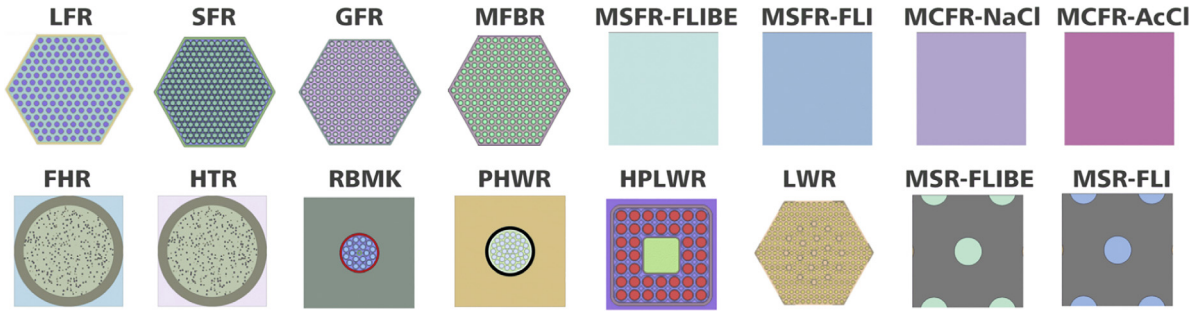

Fig. 3 Illustration of 16 simulated reactor lattices. From Krepel J and Losa E (2019) Closed U-Pu and Th-U cycle in sixteen selected reactors: Comparison of major equilibrium features. *Annals of Nuclear Energy* 128: 341–357.

Since the study is limited to lattice calculations, the multi-zone cores in this assessment are represented only by one zone. This is also the reason why only one representative zone was selected for HPLWR and why Boiling Water Reactor (BWR) was excluded. It has flooded part, which performs similarly as Pressurized Water Reactor (PWR) and voided part with much harder spectrum. Nonetheless, the parametric moderation study provides certain insight for possible BWR performance. The third most abundant reactor type, PHWR, was included in this study, but the Gas Cooled Heavy Water Reactor (GCHWR) was not. The graphite moderated light water cooled RBMK was included; however, the CO_2 cooled Advanced Gas cooled Reactor (AGR) was not included. Whereas GCHWR is not operated anymore, the AGR exclusion is not scientifically justified and it is arguably the major reactor spectrum type missing in this comparison.

Equilibrium neutron spectrum

Equilibrium neutron spectrum is one of the three characteristics of the Eigen-state (the solution of the Bateman equations). The spectrum and the equilibrium fuel composition mutually influence each other (see Fig. 1). The initial and equilibrium fuel compositions provide different spectra. For dedicated U-Pu breeders the initial fissile load consists of plutonium from irradiated LWR fuel and the difference is small. For dedicated Th-U breeders it is more complex, because the initial fissile fuel, plutonium or enriched uranium, is not extensively present in the thorium irradiation chain. The difference between initial and equilibrium spectrum is thus not negligible. Nevertheless, for the purposes of this study it is still quite small, because the spectrum is mostly shaped by scattering materials and major fertile nuclide. In reactors, initially using ^{235}U fissile fuel, the equilibrium spectrum differ. However, the differences are smaller than expected and it seems that the effect of scattering and capture cross-sections of the coolant and structural materials as well as of the fertile fuel play a dominant role in spectrum determination. Hence, the equilibrium neutron spectrum is discussed here as the first result, because it is rather spectrum which influences fuel composition than vice versa.

To understand the spectrum differences between the 16 selected reactor types and between the LWR parametric cases, each spectrum is graphically represented as a point in a 2D plot; where the x coordinate represents the relative share of thermal neutrons (0–1 eV) and the y coordinate the relative share of fast neutrons (0.1–20 MeV). The resulting plots for both the Th-U and U-Pu cycles are shown in Fig. 4. This figure is used for general spectrum discussion. For a more detailed discussion, refer to Krepel and Losa (2019).

All fast reactors included in this study have zero share of thermal neutrons in their spectra. Hence they are vertically aligned on a line x = 0 according to their share of fast neutrons. The MSFR with softest fast spectrum is the lowest point on this line. The softening is enhanced by beryllium presence. Surprisingly, the share of fast neutrons around 30–35% in MSFR spectrum is comparable to that in LWR. It seems that ^{7}Li and ^{19}F, even though not thermalizing extensively the neutrons, are slowing the neutrons out of the fast energy range due to their scattering cross-section resonances at relatively high energies. For the respective cross-sections and spectra, please refer to Fig. 2 from Krepel and Kramer (2021) and to Fig. 7 from Krepel and Losa (2019). Other fast reactors have at least 55% of fast neutrons in their spectra. The hardest spectrum with more than 70% of fast neutrons has MCFR based on the AcCl salt. There are no structural materials in this homogeneous reactor and the only non-fuel nuclide, ^{37}Cl, has quite a low scattering cross-section with narrow resonances and features a relatively high mass. Hence, it is not softening the spectrum like, for instance, ^{23}Na having lower mass and higher scattering cross-section with broader resonance. The three classical fast reactors, SFR, GFR, and LFR are located practically at the same spot. Of the fast reactors with solid fuel, only the MFBR has somewhat harder spectrum. Its fuel in metallic form, without light elements like O_2 in oxide fuel, poses higher actinides density; the heterogeneity effect is thus pronounced and Na scattering relatively weaker. The difference between the Th-U cycle and U-Pu cycle in this graphical representation is negligible for fast reactors. Only the MCFR core seems to have harder spectrum in the U-Pu cycle. It is probably related to the higher rate of direct ^{238}U fission, which reduces the overall absorption rate of actinides. For more detailed discussion, refer to Krepel and Losa (2019).

The thermal reactors included in this study are located in the bottom half of Fig. 4. They are scattered along the diagonal, because moderation simultaneously decreases the fast neutron share and increases the thermal neutron share. The thermal spectrum share ranges from 25% to 80%. The hypothetical diagonal line, connecting bottom left and upper right corners in Fig. 4, represents reactors with 20% share of neutrons in the intermediate energy range (1 eV–0.1 MeV). Close to this line are reactors with very hard spectrum as MCFR or with very soft spectrum as FHR and HTR. It seems that the majority of the neutrons in these reactors have one preferable energy range and the intermediate range is thus suppressed. Reactors using graphite, heavy water and light water as a moderator and light water as a coolant (RBMK, PHWR, LWR, and HPLWR) are almost equally distant from the diagonal

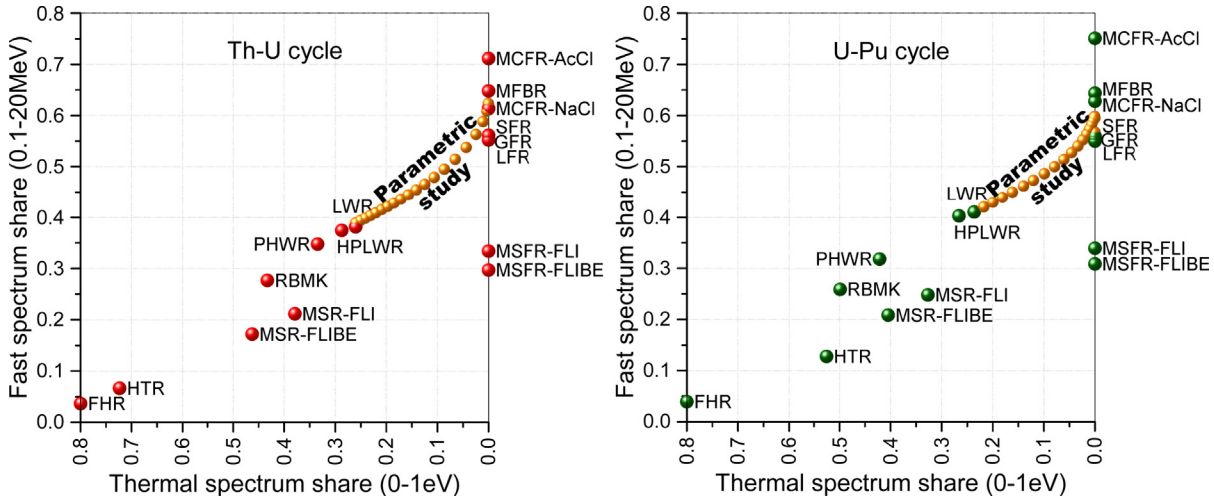

Fig. 4 Equilibrium spectra distribution in Th-U (left) and U-Pu (right) cycles.

line. The worst from this perspective are fluoride salt based MSR and MSFR. The high share of intermediate neutrons in MSR and MSFR can be caused by the above discussed features of ^{7}Li and ^{19}F. At the same time, it is codetermined by homogeneous dilution of actinides in the salt. The heterogeneity effect is thus strongly suppressed and fast neutrons fission is higher in LWR than in MSFR or MSR.

The FHR and HTR are the two reactors with the lowest specific fuel density and fuel-to-moderator ratio in this study. In addition, they are designed for uranium with enrichment between 10–20%; but the equilibrium fuel composition includes only 0.5% or 1.5% of fissile nuclides for the U-Pu or Th-U fuel cycle, respectively. The total cross-section of actinides is thus very low and more neutrons are in the thermal equilibrium with the moderator than in the slowing down process. The presence of cooling salt in FHR case introduces additional parasitic neutron captures. As a consequence, FHR is less sensitive than HTR to the remarkable difference of synthetic actinides share between the Th-U and U-Pu cycles. The FHR and MSR-FLIBE are composed from the same elements; however, they differ in salt volumetric share and actinides specific density.

The LWR parametric study practically connects the solid fuel fast reactors with the LWR point. Light water moderated reactors (LWR and HPLWR) have specific fuel density comparable to fast reactors, and the fast neutrons share in their spectra is slightly higher than the thermal neutrons share. In heavy water moderated and light water cooled PHWR the proportions are the same. The remaining reactors are moderated by graphite, and the thermal neutron share dominates. The position of these reactors in the chart depends on fuel-to-moderator ratio, where the lower specific fuel density results in softer spectrum. In the RBMK case it is partly biased by the light water coolant. In Hombourger et al. (2015) and Hombourger (2018) another parametric study was performed with EQL0D v3 for graphite moderated MSR operated in the Th-U closed fuel cycle. The results of this study practically linearly span from MSFR through MSR towards FHR. The highest reactivity in the moderated area was obtained for the 15% salt share in the core and 5 cm channel radius; the case which was adopted for the present study.

Equilibrium fuel composition

The second major feature of the equilibrium state is the fuel composition. It represents a state to which every non-equilibrium fuel composition evolves during irradiation. It also determines the waste composition from reprocessing losses. Furthermore, together with the equilibrium spectrum, it determines the equilibrium reactivity.

Sixteen reactors comparison study

The equilibrium fuel composition for each of the 16 selected reactors and for each fuel cycle is presented in **Fig. 5** using masses relative to the major fissile nuclides, i.e., ^{233}U or ^{239}Pu. Accordingly, these two nuclides are not plotted in **Fig. 5** and the related discussion refers only to other synthetic actinides. The results for both cycles in **Fig. 5** are further divided into thermal and fast reactors charts. This division is driven solely by the different scale needed for thermal reactors in the U-Pu cycle. Since the simulation assumes continuous refueling of fertile feed, all nuclide masses are constant. For each nuclide the creation and destruction rates are in balance. The reactor spectrum type determines the reaction probabilities and these probabilities dictate the equilibrium mass of each nuclide. In all fast reactor cases, the synthetic actinides share is below 1. The only exception is MSFR with the softest fast spectrum in the U-Pu cycle.

In the Th-U cycle the synthetic actinides share is close to 1 also for thermal reactors. This is actually the reason why the Th-U cycle can be considered for thermal breeder reactors. It is the consequence of ^{233}U low capture probability, which stays around 10% of total fission rate also in thermal spectrum. The respective ^{239}Pu capture probability in thermal spectrum is roughly 3x higher, refer to Krepel (2021a). Hence, also the creation rate of higher nuclides is 3x higher in the ^{238}U irradiation chain than in the ^{232}Th irradiation chain.

In the Th-U cycle, a slightly higher share of synthetic actinides can be seen for HTR and FHR. These two reactors have the highest specific power. Accordingly, rate of synthetic actinides creation is influenced by increased fraction of ^{233}Pa and its transmutation into ^{234}U. Since the ^{233}Pa amount is proportional to specific power, this effect can be seen in smaller scale also for MFBR, where the synthetic actinides share is one of the lowest between fast reactors in the U-Pu cycle, but one of the highest in the Th-U cycle. Obviously, ^{239}Np with 11x shorter half-life for radioactive decay, refer to Table 1 from Krepel (2021a), does not affect the ^{240}Pu mass in the same manner as ^{233}Pa affects the ^{234}U mass.

In the U-Pu cycle the relative synthetic actinides amount in thermal reactors ranges from 4 to 12. Due to the energy dependent ^{239}Pu capture probability, this fuel cycle is much more sensitive to spectrum variation. Accordingly, for majority of thermal reactors the equilibrium fuel composition alone (not accounting for parasitic absorptions by FPs, coolant and structural materials) has too high parasitic capture and results in negative reactivity.

The relative masses of other synthetic actinides in **Fig. 5** are useful for the neutron balance considerations (Krepel and Losa, 2019). However, for other fuel cycle features, as for instance the radiotoxicity due to reprocessing losses, the absolute share of main fissile nuclide and of other synthetic actinides are more relevant. According to **Fig. 6**, the main fissile nuclide share of all actinides is ~10% in all fast reactors and in both fuel cycles. This ratio is determined by the respective capture and fission cross-sections in the fast neutron spectrum of the main fertile and fissile nuclides, respectively. The same ratio in thermal spectrum strongly differs between the fuel cycles. It is slightly less than 1.5% in Th-U cycle and 0.5% in the U-Pu cycle. The lower ratio in the U-Pu cycle is a consequence of higher ^{239}Pu fission cross-section and its resonance in the thermal energy range.

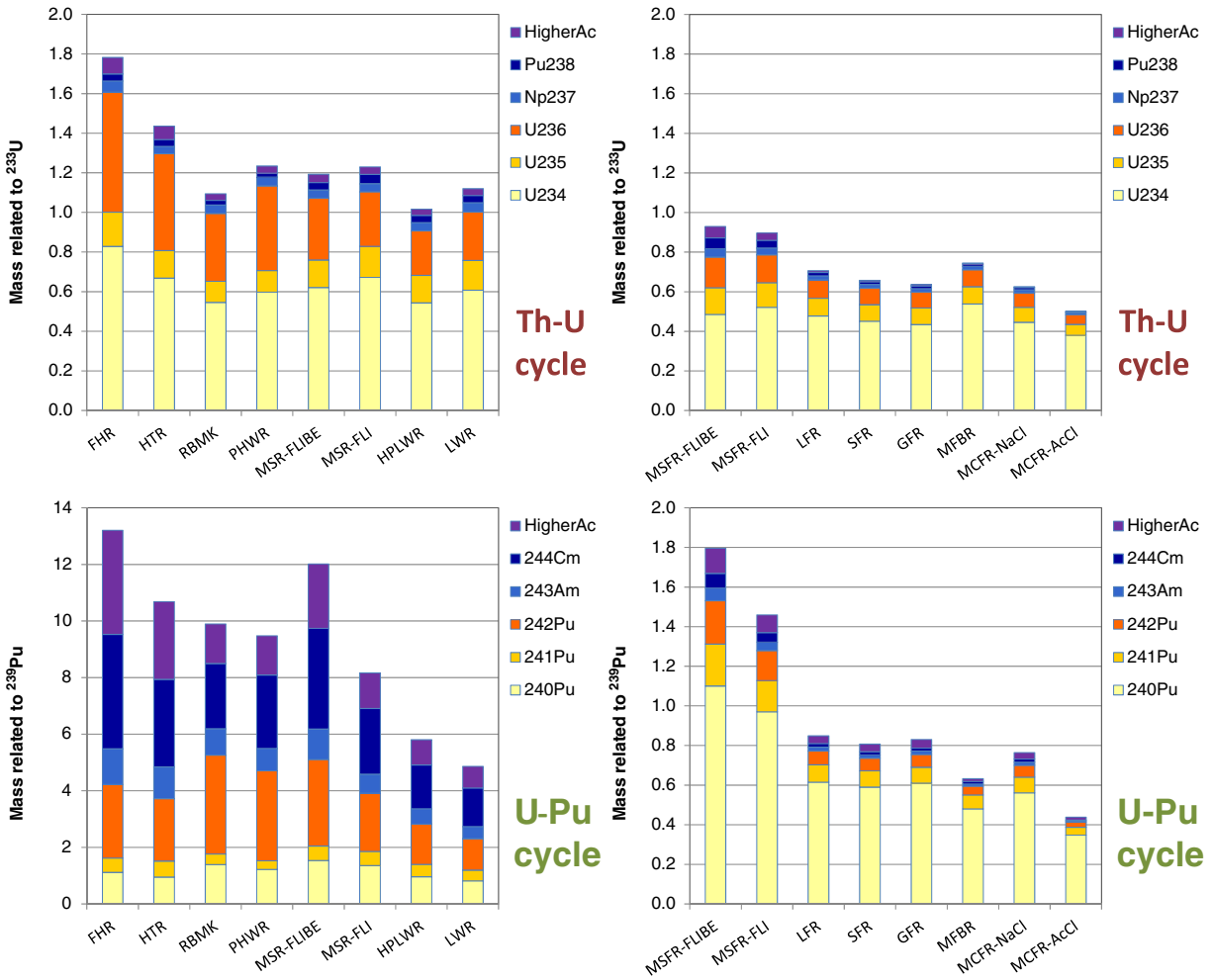

Fig. 5 Actinides mass related to the main fissile nuclide in Th-U (top) and U-Pu (bottom) cycles. From Krepel J and Losa E (2019) Closed U-Pu and Th-U cycle in sixteen selected reactors: Comparison of major equilibrium features. *Annals of Nuclear Energy* 128: 341–357.

The actinides parasitic neutron capture usually does not allow for breeding in thermal spectrum. Furthermore, these reactors are very sensitive to FPs accumulation (Krepel, 2021b). This is valid especially in the U-Pu fuel cycle, where the fissile actinides share is 3 times lower than in the Th-U fuel cycle. The share of fissile nuclides is given by the respective cross-sections, which are actually strongly co-determined by the neutron pairing effect. The ^{233}U and ^{239}Pu nuclei with odd neutrons are more eager to interact with neutrons (have bigger cross-sections and bigger fission chance) than ^{232}Th and ^{238}U nuclei with even neutrons (Krepel, 2021a).

Fig. 6 also shows that the absolute amount of other synthetic actinides is much lower in thermal spectrum, around 1.8% in the Th-U and 2.5% in the U-Pu fuel cycle. Only HTR and FHR, as the outliers, have somewhat higher synthetic actinides share, 2.5–3.5% in the Th-U cycle and 5.5% in the U-Pu cycle. For fast rectors, MSFR-FLI and MSFR-FLIBE are the outliers. The second named has the highest synthetic actinides share of 10% and 18% in the Th-U and U-Pu cycle, respectively. In other fast reactors this share is around 7% in the Th-U and 9% in the U-Pu cycle. Only the MCFR-AcCl spectrum is sufficiently hard so that synthetic actinides share of around 5% is the same in both fuel cycles. In all other cases the Th-U cycle feature a smaller synthetic actinides share than the U-Pu cycle. In general, fast reactors have 4x higher absolute share of synthetic actinides share than thermal reactors. Accordingly, should the same absolute share of legacy actinides be added to the equilibrium composition in all reactors, thermal cores will transmute it up to four times faster (Krepel and Losa, 2016). However, as Fig. 5 or (Krepel, 2021b) indicates, it will cost more neutrons in the thermal spectrum.

The synthetic actinides share in the fuel determines their amount in recycling losses and together with the half-lives also the radiotoxicity. The share of synthetic actinides in both fuel cycles is up to four times lower in thermal reactors, and in all reactors it is also smaller for the Th-U cycle. Furthermore, this cycle profits from the fact that the synthetic actinides have longer half-lives than the respective nuclides in the U-Pu cycle. Thus the radiotoxicity is typically lower at the early stage for the Th-U cycle. However, as a consequence of the longer half-lives, a secondary peak may occur for the Th-U cycle and the U-Pu cycle may provide lower radiotoxicity in the longer term (Krepel, 2021a).

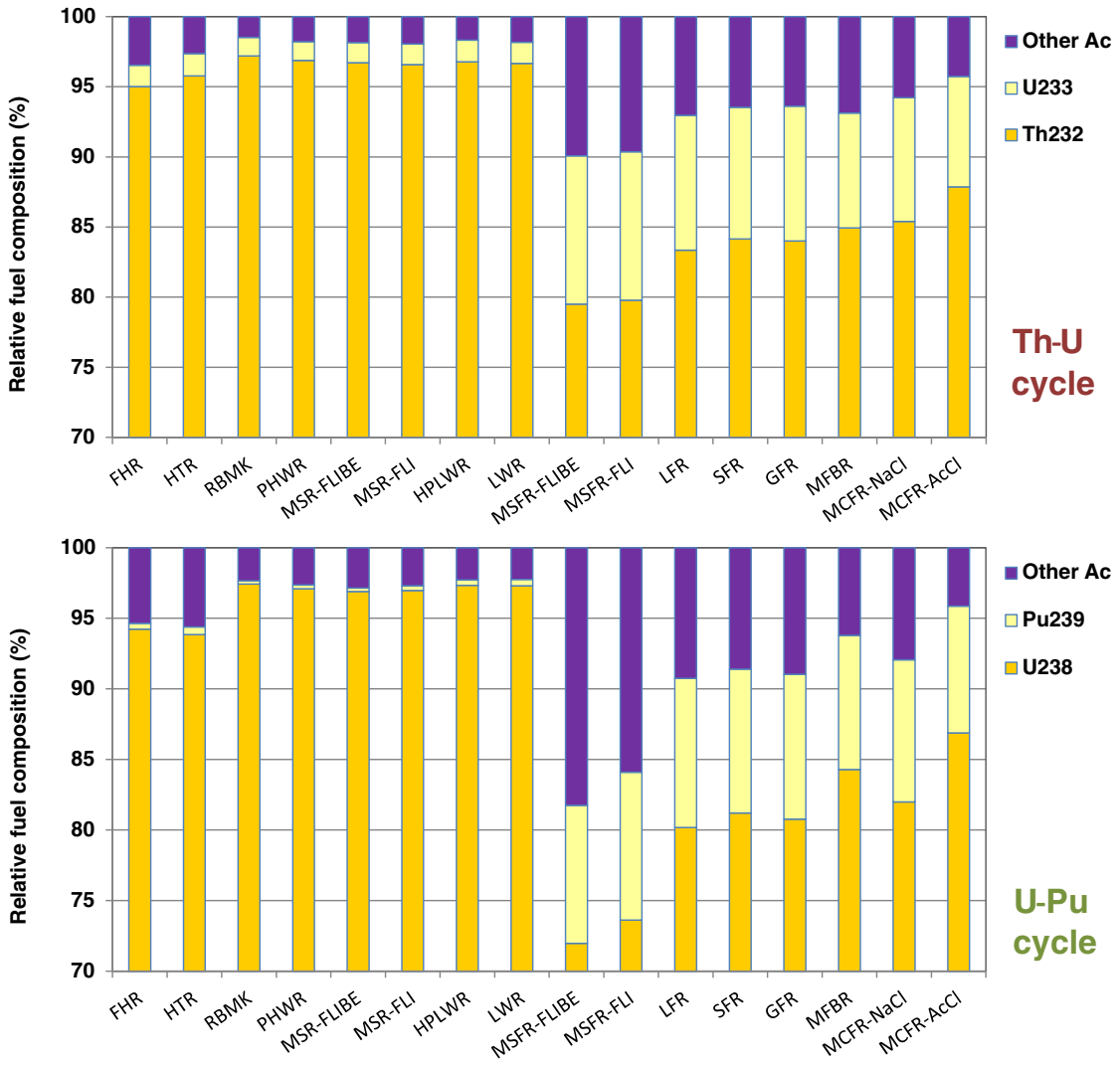

Fig. 6 Actinides relative composition in Th-U (top) and U-Pu (bottom) cycles.

LWR parametric moderation study

The high fission-to-capture probability of ^{233}U over a wide energy range in the Th-U cycle is well illustrated by the LWR moderation study. In this study, the water moderator is step-by-step removed from the reactor. The synthetic actinides share related to the ^{233}U mass stays constant at 1.1 level (see top Fig. 7) up to about 30% moderator density. From about 20% moderation, with water being almost completely removed, and the neutron spectrum getting very hard, the synthetic actinides share related to ^{233}U drops down to 0.6. In the U-Pu cycle, the variable ^{239}Pu fission-to-capture probability causes gradual drop of the ratio between synthetic actinides and the main fissile nuclide ^{239}Pu from 5 at full moderation to 1.1 at 50% moderation. Then there is a plateau, or even slight increase, and only the unmoderated case (0% water density) drops below this ratio being equal to unity (see bottom Fig. 7).

The relative masses in Fig. 7 mirror the fission-to-capture probability of ^{233}U and ^{239}Pu. The absolute shares of main fissile nuclides and of other synthetic actinides, presented in Fig. 8, are rather determined by the respective capture and fission cross-sections of the main fertile and fissile nuclides. The variation of fissile share with moderation has a similar tendency for both the Th-U and U-Pu fuel cycles, but it is not the same. The share of main fissile nuclide is $\sim$10% in fast spectrum (0% moderation) for both cycles. It drops to 3% at 25–30% moderator density in the Th-U cycle, and at 40–45% moderator density in the U-Pu cycle. At 100% moderation the share is around 1.5% in Th-U cycle and 0.5% in the U-Pu cycle. In the thermal spectrum (70–100% moderation), the total amount of synthetic actinides is the same for both cycles, or even slightly lower for the U-Pu cycle. However, the split between the main fissile and the other synthetic actinides is different in the two fuel cycles.

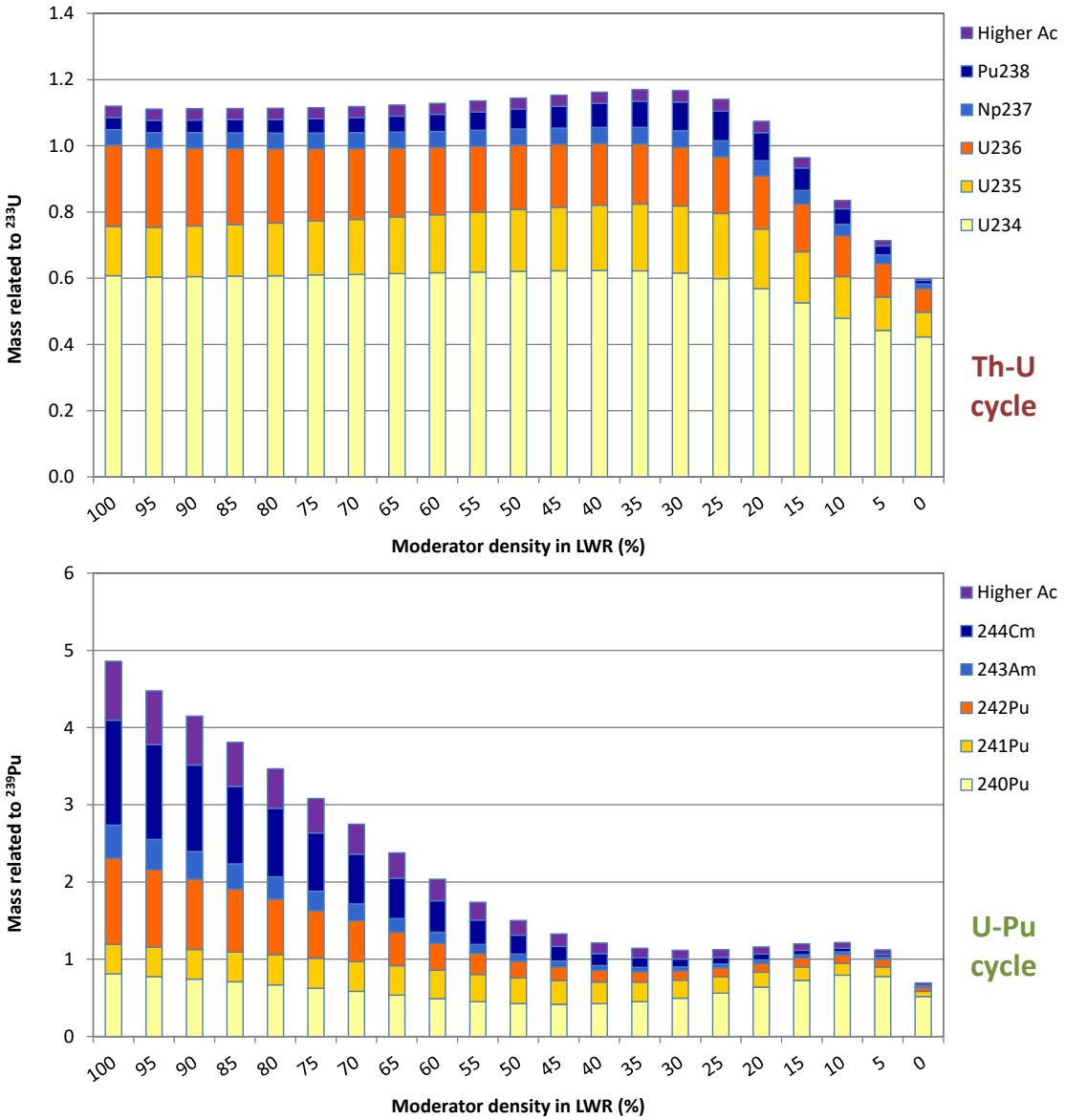

Fig. 7 Actinides mass related to the main fissile nuclide in Th-U (top) and U-Pu (bottom) cycles for the LWR parametric moderation study.

Equilibrium multiplication factor

The equilibrium multiplication factor is the major indicator for the neutron economy. Since the fuel composition is stabilized and does not evolve during irradiation, all simulated reactors act as self-sustaining breeders. Nonetheless, not all reactor can reach criticality with the equilibrium fuel composition. Accordingly, the respective equilibrium reactivity is indication of the capability of the reactor to be operated as a self-sustaining breeder. In Krepel (2021b) the reactors are classified based on this reactivity.

The infinite multiplication factor for the 16 simulated reactors follows the expected trend. Since the reactors are roughly sorted in Fig. 9 by their spectrum, for both fuel cycles, there is an overall increase in slope from left to right as the reactivity grows with spectrum hardening. The increase is higher for the U-Pu cycle, due to its higher sensitivity to spectrum variation and its effect on synthetic actinides build up (see Figs. 5 and 7). The trend in Fig. 9 is, however, perturbed by varying parasitic neutron captures of coolants and structural materials.

As expected, practically all thermal reactors do not have sufficient equilibrium reactivity to be operated as self-sustaining breeders. For the Th-U cycle, in which the synthetic actinides mass stays low even in thermal spectra, there are few thermal reactors with positive or close to zero reactivity. These reactors are moderated by low capture moderators like heavy water or graphite. However, it should be recalled that the values of neutron multiplication factors in Fig. 9 were derived from infinite lattice calculations and with the neglect of FPs. Accordingly, the excess reactivity will be lower in reality. Potentially, also beryllium could act as

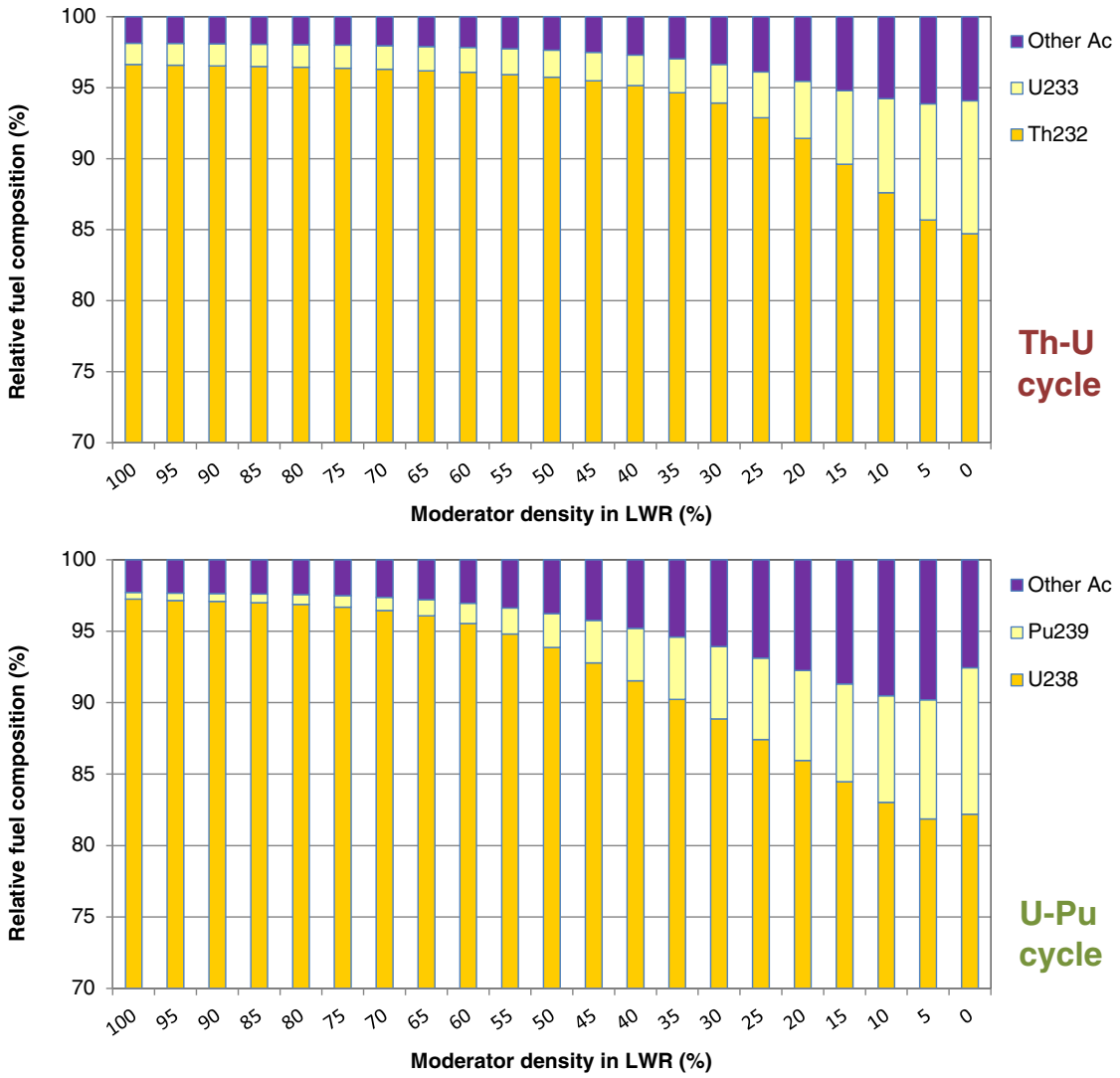

Fig. 8 Actinides relative composition in Th-U (top) and U-Pu (bottom) cycles for the LWR parametric moderation study.

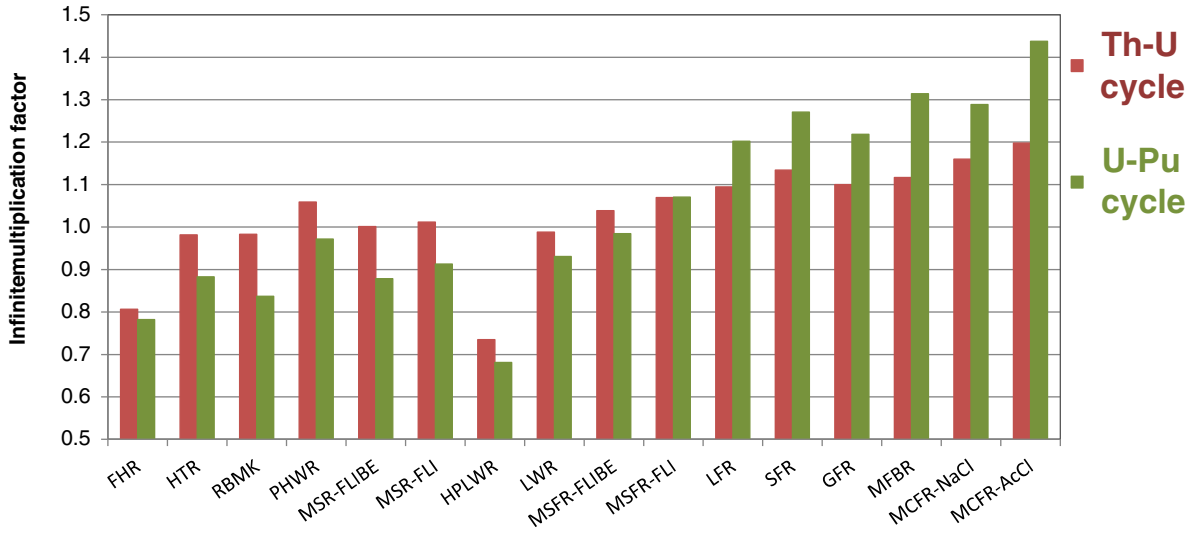

Fig. 9 Equilibrium multiplication factor for Th-U and U-Pu cycles and 16 selected reactors.

low absorbing moderator, e.g., for MSR. Nonetheless, both heavy water and beryllium needs to be separated from the MSR fuel by structural materials, which may deteriorate the performance (Hombourger, 2018). Furthermore, even if breeding would be possible in thermal reactors, their fissile share (see Fig. 6) is small. Hence, they will be very sensitive to FPs buildup (Krepel, 2021b). It may result in prohibitively high fuel reprocessing frequency. The only reasonable option could be the MSR, in which the FPs can be continuously extracted from the liquid fuel. However, even in this case the need of fast FPs removal may prove being prohibitive.

In the thermal spectrum cases, there are two outlier systems, FHR and HPLWR, with very low multiplication factors. In HPLWR case it is caused by parasitic neutron capture of the stainless steel cladding, which is not suitable for thermal spectra. In FHR it is due to the interplay between the parasitic neutron capture of the fluoride salt coolant, its higher volumetric share in the core, and the very low specific fuel density. The same salt in MSR-FLIBE, with higher specific fuel density and lower salt volumetric share, provides reasonable multiplication factors.

The Th-U cycle performs better than the U-Pu cycle in thermal, and worse in fast neutron spectra. The turning point is the MSFR-FLI, where the performance is the same in both cycles. It features a spectrum, in which the lower average number of neutrons per fission and the lower synthetic actinides concentration in the Th-U cycle provide similar reactivity as the higher average number of neutrons per fission and the higher synthetic actinides concentration in the U-Pu cycle. The detailed decomposition and analysis of the Fig. 9 results is presented in Krepel and Losa (2019).

The LWR parametric study provides insight for the equilibrium reactivity transition between thermal and fast reactors. Fig. 10 confirms that the reactivity changes with spectrum variation are stronger for the U-Pu cycle. At the same time the Th-U cycle is very close to $k = 1$ even for nominal water density and become critical already around 80% of the moderator density. However, for water moderator densities below 30%, the reactivity increase in the U-Pu cycle is steeper, yielding reasonable high reactivity earlier than in the Th-U cycle. Fig. 10 also illustrates the option to breed in reactor cores cooled by steam or in the upper part of BWR cores in which the exit water density is around 5% of the nominal LWR water density. It may be possible to design a water-cooled breeder (Huang and Zang, 2021) using water densities between 35% and 5% of the nominal LWR value. Optionally, a breeder with water density below 5% can be proposed solely for the heat production. Since the required steam pressure and accordingly the temperature would be lower, also the heat exchange and maximal power density would be limited.

Core size estimate

By means of Fermi's theory of the bare thermal reactor the critical core size can be estimated from the knowledge of equilibrium multiplication factor and material properties of each reactor (Losa, 2016; Krepel and Losa, 2017). The latter are a measure of how transparent the reactor core is for the neutrons. Application of this theory provides only very rough estimate, because the impact of neutron reflector or blanket is not accounted for. At the same time, it is based on an overestimated equilibrium multiplication factor, with neglected FPs parasitic absorption. Accordingly, it can provide certain insights for the interplay between equilibrium reactivity and reactor transparency for the neutrons.

The estimated core radii of bare self-sustaining breeders are presented in Fig. 11. The graphite moderated MSRs have quite substantial core radius. To obtain realistic core size, blanket and/or reflector should surround the core and the FPs must be continuously removed from the core. Furthermore, the neutron balance should be improved by ^{233}Pa extraction from the molten salt. Its radioactive decay outside of the neutron flux will increase the ^{233}U and reduce the ^{234}U production rates (Krepel, 2021a).

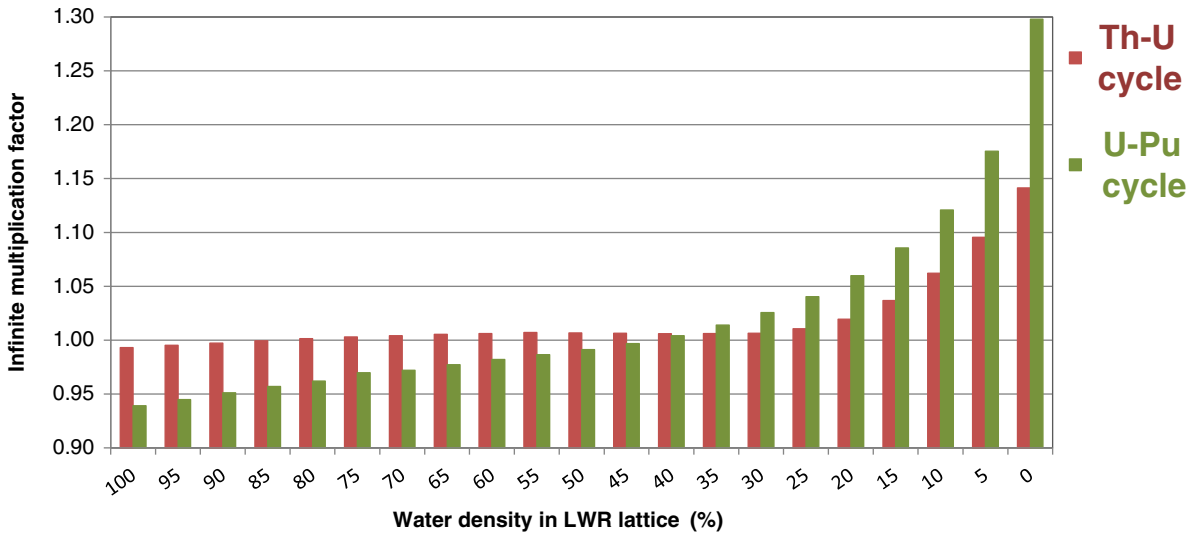

Fig. 10 Equilibrium multiplication factor for Th-U and U-Pu cycles and the LWR parametric moderation study.

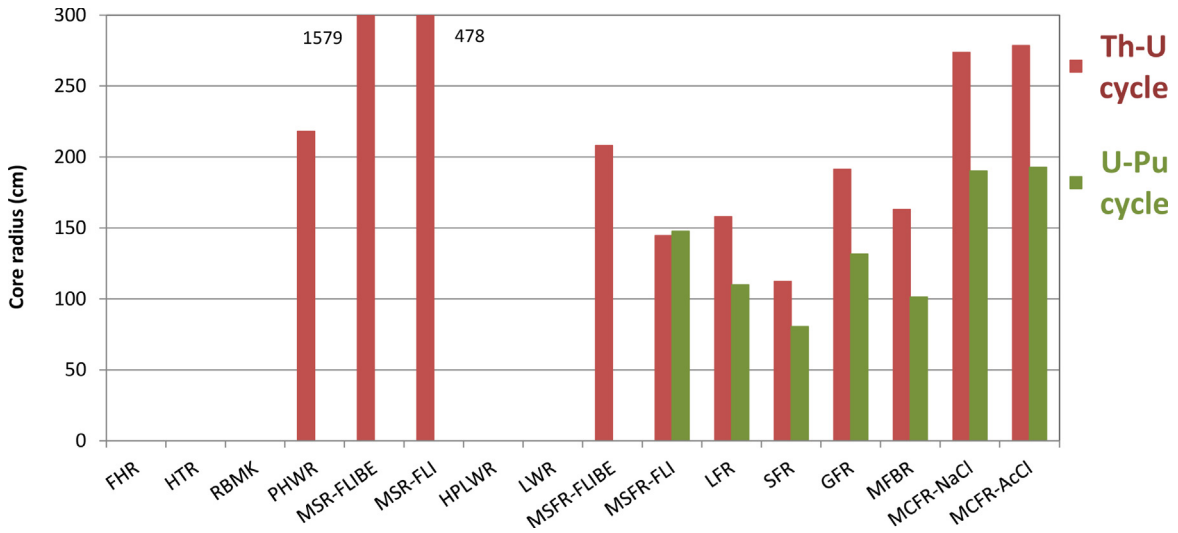

Fig. 11 Estimate of bare critical core radius for Th-U and U-Pu isobreeding fuel cycles.

Optionally, some of the synthetic actinides (e.g., ^{237}Np) may not be recycled to decrease their contribution to parasitic neutron captures. The PHWR displays a smaller core radius than the MSR, but it has a solid fuel. Continuous FPs separation and ^{233}Pa removal is thus not possible. The fuel reprocessing frequency would need to be quite high to minimize the FPs captures and achieve breeding.

The SFR seems to provide the best combination of equilibrium multiplication factor and core material properties for a compact breeder. Especially in the U-Pu cycle the core is quite small. On the other hand, the MCFR represents an example of imbalance between the parameters. It has the highest equilibrium multiplication factor (see Fig. 9) and (together with the HTR and FHR) is the most transparent core for the neutrons. As a result, the core radius of the self-sustaining MCFR breeder is the largest of the fast reactors.

The estimate of minimum core radius for the LWR parametric study shows an interesting trend. For the Th-U cycle there are two minima, one at 55% moderation and the second at 0% moderation. The equilibrium multiplication factor between 100% and 30% moderation in Fig. 10 is almost constant, with very tiny local maxima at 55% moderation; nonetheless, the core radius is very sensitive to the moderation values (Fig. 12). The second minimum at 0% is due to the steep increase in multiplication factor as water density decreases below ~20%. However, the core radius at 55% moderation is still too big for practical application. Interesting cases with smaller core radius <200 cm are obtained for the U-Pu cycle in the range of 20–0% moderation and for the Th-U cycle in the range of 5–0% moderation (which is, probably, not practical from the heat removal perspective).

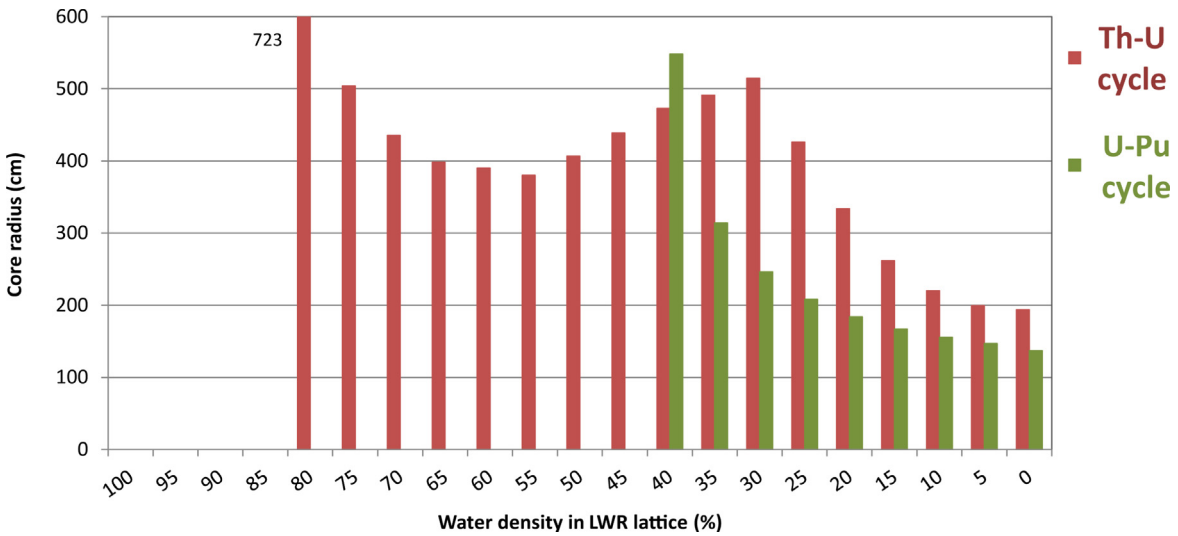

Fig. 12 Estimate of bare core radius for Th-U and U-Pu isobreeding fuel cycles in the LWR moderation parametric study.

Equilibrium temperature coefficients

The equilibrium state can be evaluated also from a safety perspective. In this article, only the fuel and coolant temperature coefficients of reactivity have been evaluated. To obtain these coefficients, the respective material properties are perturbed as follows: temperature of the fuel is increased by 300 °C at constant density, and density of the coolant is reduced by 10% at constant temperature. This approach allows for separation of the temperature and density effects for cases with fluid fuel.

Fluid density change

In all cores, the fluid density reduction results in decrease of the fluid capture and scattering probabilities. Compared to other core materials, its relative importance is thus reduced. The only exceptions are the homogeneous MSFR and MCFR, where the core is exclusively filled by the fluid (that functions as both fuel and coolant). In this case, the fluid density change represents a global density reduction, and since an infinite lattice is simulated, it has no impact on the reactivity (results absent in **Fig. 13**). In reality fluid density reduction will increase neutron leakage from a finite core and provide a negative reactivity effect.

The reduced capture probability results always in positive reactivity. The reduced scattering probability induces spectrum hardening that can introduce positive or negative reactivity. In fast reactors it introduces a positive reactivity (Krepel et al., 2011). It reduces the neutron capture rate in the resonance area and it increases the fission rate above the resonance area. The induced positive reactivity is higher in the U-Pu cycle (see LFR, SFR, GFR, and MFBR in **Fig. 12**), because the slope of the respective fission cross-section is steeper with increasing neutron energy as shown by bottom Fig. 17 from Krepel and Losa (2019) and because the resonances of the capture cross-section start at higher energies in the U-Pu cycle (Shusterman, 2021). Consequently, the positive coolant density effect for fast reactors is stronger in the U-Pu cycle (Fiorina et al., 2013b).

In thermal reactors, where the fluid also moderates the neutrons, the reduced scattering probability introduces negative reactivity. Interplay between this negative reactivity and positive reactivity introduced by capture probability reduction is crucial for the safety of moderated reactors. The overall effect should be negative. Hence, moderated cores are usually designed to be under-moderated, so that the moderator density reduction results in negative reactivity as for instance in PHWR and LWR. In many thermal reactors: FHR, HTR, RBMK, PHWR and MSR, dedicated coolant fluid is used, which has only minor moderation function. Its density effect is thus dominated by the capture component and results in positive reactivity. For PHWR coolant the capture component only slightly dominates above the scattering component and the effect is slightly positive. Nonetheless, in reality the increased leakage rate will somewhat compensate for the reduced capture rate. In MSR the fluid includes fuel and its destiny reduction reduces the fuel-to-moderator ratio (Krepel et al., 2014). In some cores there are two fluids with separate moderation and cooling functions. For such cores two independent results are presented in **Fig. 13**.

In the HPLWR the results differ qualitatively between the Th-U and U-Pu cycles. However, a big difference between the fuel cycles can be seen also for FHR, PHWR, MSR-FLI and LWR. In the U-Pu cycle, ^{239}Pu fission cross-section has a large resonance at 0.3 eV. A similar but weaker resonance of ^{233}U fission cross-section in the Th-U cycle is located at 1.8 eV, as indicated by top Fig. 17 from Krepel and Losa (2019) and Shusterman (2021). Nonetheless, the ^{239}Pu resonance has higher amplitude, it is broader, preceded by cross-section suppression, and followed by larger cross-section than in the ^{233}U case. Accordingly, in the U-Pu cycle the negative moderation effect is stronger. This can be seen particularly by HPLWR, where the moderator is located in a dedicated central tube (see **Fig. 3**) and its density effect is dominated by capture rate in the Th-U cycle and the moderation effect in the U-Pu cycle. In

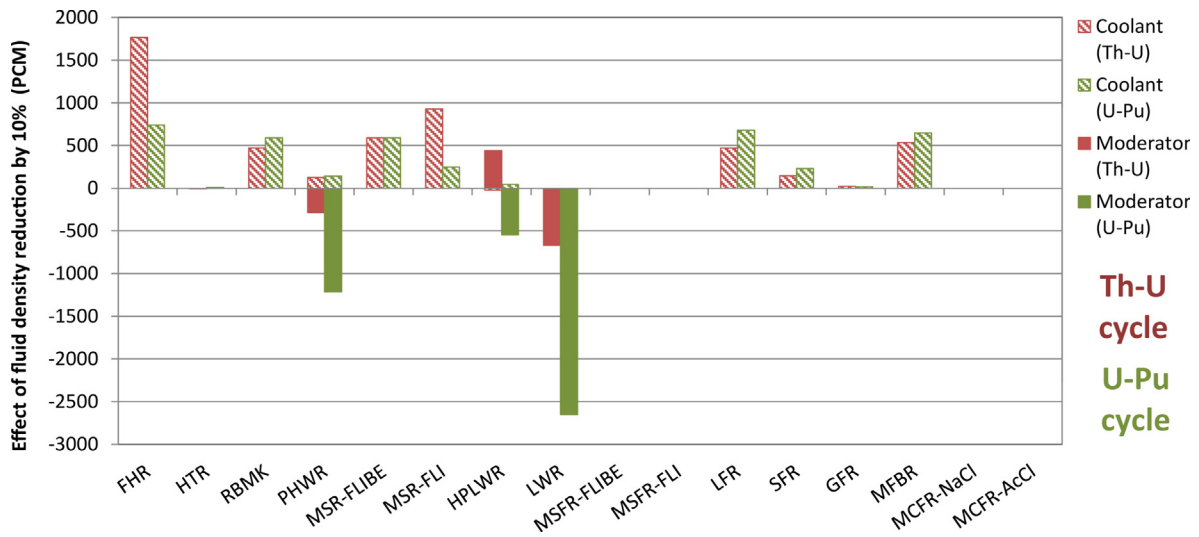

Fig. 13 Reactivity induced by 10% reduction of the fluid density for Th-U and U-Pu cycles. In PHWR and HPLWR cases the moderator and coolant densities were reduced independently. From Krepel J and Losa E (2019) Closed U-Pu and Th-U cycle in sixteen selected reactors: Comparison of major equilibrium features. *Annals of Nuclear Energy* 128: 341–357.

PHWR the coolant and moderator density effects have opposite sign. It seems that the reactor is under-moderated from moderator perspective and over moderated from coolant perspective.

The results for LWR moderation study shown in Fig. 14 combine all above discussed effects. At 0% moderation there is no fluid in the core and the respective fluid density effect is zero. In thermal spectrum the negative reactivity effect is much stronger in the U-Pu cycle. This effect is primarily dictated by the ^{239}Pu fission cross-section characteristics that is also responsible for the relatively small ^{239}Pu fraction in Fig. 8. To a certain extend there is a correlation in the thermal region (100–65% of nominal water density)—the bigger the fissile share presented in Fig. 8, the smaller is the fluid density effect in Fig. 14. Higher fissile share implies a lower fraction of the fissions in the thermal energy range. It becomes unimportant at moderation levels corresponding to 45–5% of nominal water density. In this region the U-Pu cycle features less negative and eventually positive coolant density effect, because of the enhanced fission probability induced by spectrum hardening.

Doppler effect

The fuel temperature increase results in so called Doppler effect, or actually broadening of cross-section resonances. Since the cross-section resonances of the fertile fuel dominate neutron capture, the Doppler effect always results in negative reactivity. The only exception, PHWR in the U-Pu cycle, will be discussed separately. Since the respective mechanisms differ between fast and thermal reactors, the absolute reactivity change is presented in Fig. 15.

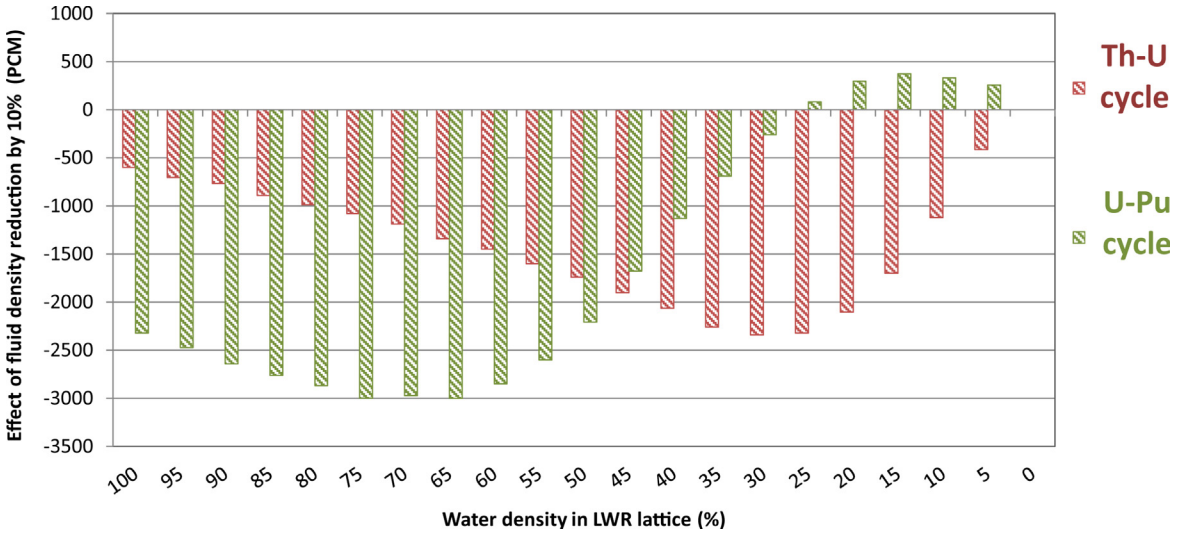

Fig. 14 Reactivity induced by 10% reduction of the water density in the LWR parametric moderation study for Th-U and U-Pu cycles.

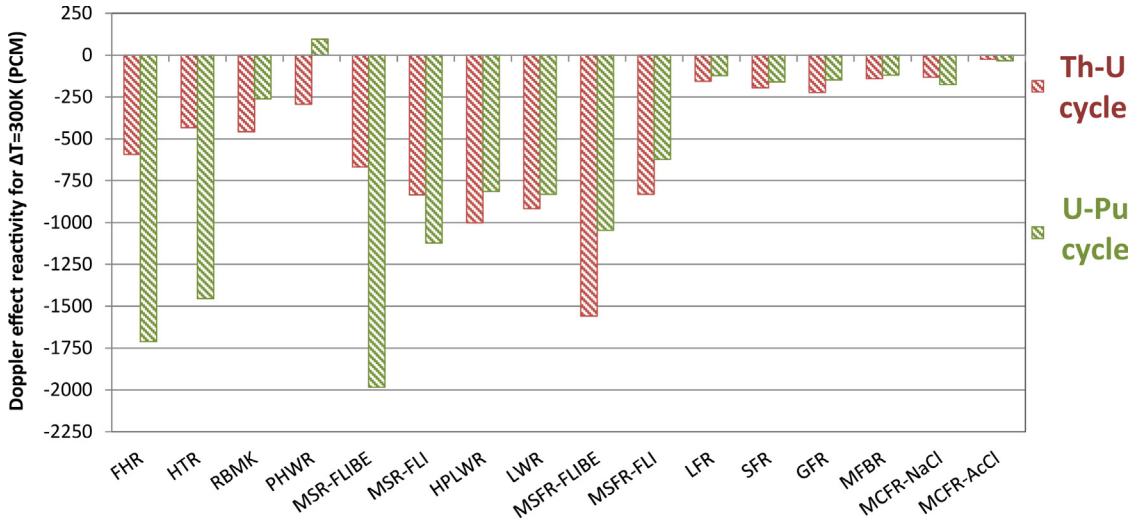

Fig. 15 Doppler reactivity effect induced by 300 °C fuel heat-up in Th-U and U-Pu cycles. From Krepel J and Losa E (2019) Closed U-Pu and Th-U cycle in sixteen selected reactors: Comparison of major equilibrium features. *Annals of Nuclear Energy* 128: 341–357.

In fast reactors only the low-energy tail of the neutron spectrum is in the resonance area. Moreover, the ^{232}Th resonances start later at lower energies than for ^{238}U. The Doppler effect in fast reactors is, in simplified way, composed of negative reactivity caused by the enhanced capture in resonances and of positive reactivity from spectrum hardening induced by this enhancement (Krepel et al., 2011). Since the U-Pu cycle profits more from spectrum hardening, its resulting Doppler effect is less negative in fast reactors. The only exception is the MCFR core which features the hardest spectrum. In this reactor there are almost no neutrons in the energy range of the ^{232}Th resonances so the respective Doppler effect is smaller than in the U-Pu fueled reactors.

In thermal reactors the Doppler effect reduces the neutrons resonance escape probability during the slowing-down process resulting in a reduction of thermal spectrum component. Both the increased resonance capture rate and the suppression of thermal spectrum result in negative reactivity. The magnitude of the Doppler effect in thermal reactors depends on the moderator properties and on the core heterogeneity. The latter effect is strong in all water moderated or cooled reactors including PHWR and RBMK. In these reactors the Doppler effect has medium amplitude and is stronger for the Th-U cycle. In reactors with very weak heterogeneity (FHR, HTR and MSR) the resonance escape probability for the neutrons during the slowing-down process plays much more important role and the respective Doppler effect is very strong especially for the U-Pu cycle. Nonetheless, it is rather a complex phenomenon which is also effected by the concentration of other synthetic actinides in the core.

The U-Pu cycle in PHWR core is an exception. It features a positive Doppler effect because the dedicated heavy-water moderator does not have cooling function and its temperature is kept low. The respective Maxwellian spectrum peaks at the lowest energy, see Fig. 7 from Krepel and Losa (2019). The large resonance of ^{239}Pu fission cross-section is close to this peak. At the same time, the heterogeneity effect is very strong in PHWR and the resonance broadening in epithermal area has thus a lower impact on neutron capture. As a result, the fission resonance broadening dominate in this core.

The variation of the Doppler effect with the water density for LWR parametric study is shown in **Fig. 16**. The dominant effect is resonance capture probability that peaks at intermediate water densities which feature epithermal neutron spectrum. For very hard spectrum, the Doppler effect is the weakest due to very small resonance capture probability. It is the most negative for the Th-U cycle around 25% water density because spectrum hardening has a smaller positive reactivity effect than for the U-Pu fuel cycle. For the Th-U cycle it is the most negative around 60–75% water density because of the ^{239}Pu fission cross-section characteristics.

Summary

To fully utilize natural resources in nuclear fission reactors, self-sustaining breeding in closed cycle is necessary. In this cycle the synthetic fissile nuclides ^{233}U and ^{239}Pu acts as kind of catalyzer for energy production from primordial ^{232}Th and ^{238}U nuclides. The repetitive fuel irradiation in a closed cycle results in the equilibrium fuel composition. When only the ^{232}Th or ^{238}U primordial nuclides are used as a feed fuel, the respective equilibrium fuel composition corresponds to the ^{232}Th or ^{238}U irradiation chain. In this article the basic features of the ^{232}Th and ^{238}U irradiation chains were compared for 16 selected reactors and certain insight was provided, why some of the advanced reactors are not capable to sustain the breeding process.

EQL0D v2 MATLAB script coupled to the SERPENT 2 code was used to obtain the equilibrium state by iterative procedure on an infinite lattice. The specific power was fixed at nominal value, and the irradiation of the actinides was independent from the multiplication factor. Hence, the equilibrium state was obtained also for subcritical reactors. Since the fuel composition slightly oscillates during different phases of the closed fuel cycle, one simplifying assumption was applied to obtain the constant equilibrium vector. The FPs were neglected and each fission directly resulted in addition of ^{232}Th or ^{238}U atom. With this assumption the results become

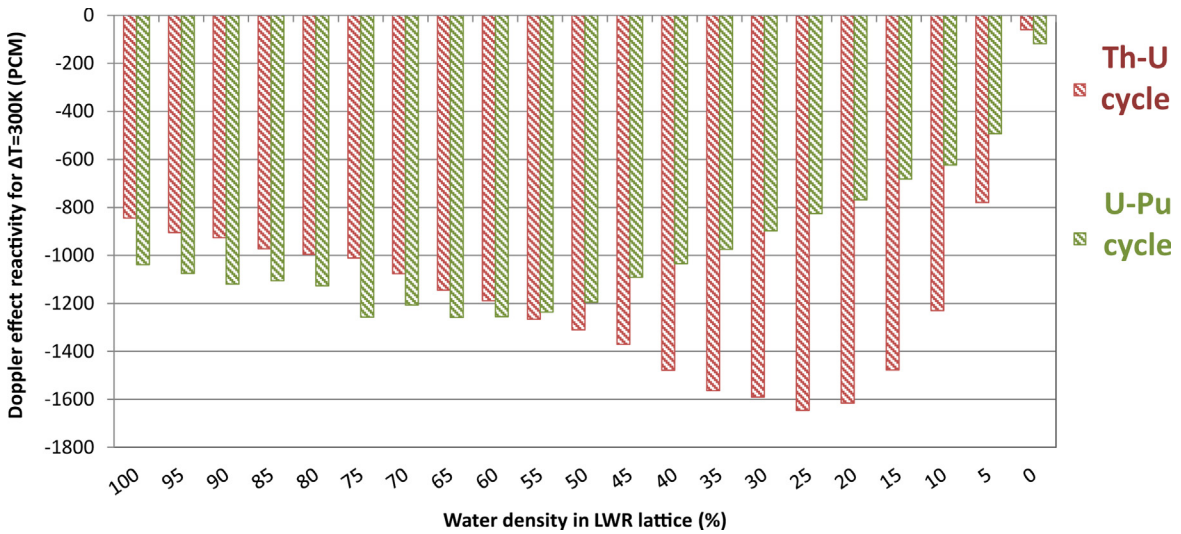

Fig. 16 Doppler reactivity effect induced by 300 °C fuel heat-up in the LWR parametric moderation study for Th-U and U-Pu cycles.

insensitive to the discharged fuel cooling time, irradiation time, reprocessing intensity and reprocessing losses. Hence, the equilibrium state depends only on the reactor specific power and core lattice geometry, composition, and temperature. In spite of the simplifications, the obtained equilibrium reactivities and actinides eigenvectors are representative for the respective reactor and many useful conclusions can be drawn.

Three major equilibrium properties were compared: excess reactivity, fuel composition, and neutron spectrum. The reactor types included in this study were selected so that all major spectrum types are covered. A LWR parametric moderation study was included to illustrate the evolution of equilibrium parameters between thermal and fast spectra. Even though the majority of the presented results is well known, this article offers a slightly different perspective for their interpretation. Moreover, the LWR parametric study provides valuable insights into the evolution of the equilibrium properties between thermal and fast reactors. Self-sustaining breeding is generally possible in fast reactors. In thermal reactors only the Th-U cycle provides positive reactivity, which, however, may not be sufficient.

The assessment of critical core radii, included in this article, is only indicative, because the FPs as well as blankets or reflectors are neglected. At the same time, this assessment nicely illustrates that self-sustaining breeding in closed cycle is nearly impossible in thermal reactors. Their neutron economy is so tight, that they can afford only negligible leakage. The resulting cores are thus very large. Furthermore, to minimize neutron losses due to FPs, prohibitively high fuel reprocessing frequency may be needed.

Among the fast reactors, the SFR seems to provide the best combination of equilibrium multiplication factor and core material properties for compact breeder. Especially in the U-Pu cycle the core is quite small. In contrary, MCFR represents an example of imbalance between the parameters. It has the highest equilibrium multiplication factor, on the one hand side, and it is the most transparent core for the neutrons, on the other. As a result, the core radius of self-sustaining MCFR breeder is the largest of the fast reactors.

Two safety related parameters, i.e., the fluid density and the fuel Doppler reactivity effects, have been evaluated. Both these effects compose of two phenomena. The fluid density effect consists of reduced fluid absorption and of spectral hardening caused by the reduced neutron scattering. The Doppler effect consists of increased absorption in resonances and of the resulting spectral shift. These effects have different amplitude and direction in different reactors.

The general conclusion of this article is that self-sustaining breeding in closed cycle can be achieved with both primordial nuclides ^{232}Th and ^{238}U practically in all fast reactors. The U-Pu cycle profits more from neutron spectrum hardening and outperforms the Th-U cycle in fast spectra. The self-sustaining breeding in closed cycle is nearly impossible in thermal reactors. Due to the lower ^{239}Pu fission probability in thermal reactors, the creation rate of higher synthetic actinides is roughly 3 times higher in the U-Pu cycle and the equilibrium fuel composition alone has prohibitively high parasitic capture. The respective fission probability of ^{233}U in the Th-U cycle is high in all spectra. It may allow for self-sustaining breeding in closed cycle in some particular thermal reactors.

References

Alemberti, A., 2021. Lead Cooled Fast Reactors (LCFR). In: Encyclopedia of Nuclear Energy, vol. 1, pp. 518–539.

Artioli, C., et al., 2009. ELSY Neutronic Analysis by deterministic and Monte Carlo methods: An innovative concept for the control rod systems. In: International Congress on Advances in Nuclear Power Plants, ICAPP'09, Tokyo, Japan.

Aufiero, M., et al., 2013. An extended version of the SERPENT-2 code to investigate fuel burn-up and core material evolution of the Molten Salt Fast Reactor. Journal of Nuclear Materials 441 (1), 473–486.

Bateman, H., 1910. The solution of a system of differential equations occurring in the theory of radioactive transformations. Proceedings of the Cambridge Philosophical Society 15 (Pt V), 423–427.

Bosq, J.C., et al., 2006. Fine 3D neutronic characterization of a gas-cooled fast reactor based on plate-type subassemblies. In: PHYSOR-2006, ANS Topical Meeting on Reactor Physics, Canadian Nuclear Society, September 2006, Vancouver, BC, Canada.

Brown, J.A., 2021. Pressurized Water Reactors. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 196–213.

Buiron, L., et al., 2007. Innovative core design for generation IV sodium-cooled fast reactors. In: ICAPP'07, Nice, France.

Chadwick, M.B., et al., 2006. ENDF/B-VII.0: Next generation evaluated nuclear data library for nuclear science and technology. Nuclear Data Sheets 107 (12), 2931–3060.

Desyatnik, V.N., et al., 1975. Density, surface tension, and viscosity of uranium trichloride-sodium chloride melts. Atomic Energy 39 (1), 649–651.

Emmett, M.B., 2000. Calculational benchmark problems for VVER-1000 mixed oxide fuel cycle. In: ANS2000 Radiation Protection and Shielding Division Topical Conference, Spokane, Washington, USA.

Fiorina, C., et al., 2013a. Investigation of the MSFR core physics and fuel cycle characteristics. Progress in Nuclear Energy 68, 153–168.

Fiorina, C., et al., 2013b. Analysis of void reactivity and Doppler coefficient for thorium- and uranium-fuelled lead fast reactor. In: ICAPP13 Conference, Jeju, Korea, April 14–18.

Fratoni, M., Greenspan, E., 2007. Determination of the equilibrium composition of cores with continuous fuel feed and removal using MOCUP. In: Proceedings of the International Topical Meeting on Mathematics and Computations and Supercomputing in Nuclear Applications, M&C + SNA-2007, Monterey, CA.

Fratoni, M., Greenspan, E., 2010. Search for equilibrium core composition methodologies for pebble bed reactors. Nuclear Science and Engineering 166, 1–16.

Fratoni, M., Greenspan, E., Peterson, P.F., 2007. Neutronic and depletion analysis of the Pb-AHTR. In: GLOBAL 2007, Advanced Nuclear Fuel Cycles and Systems, Boise, Idaho, USA.

Fütterer, M., 2021. Very High Temperature Gas Cooled Reactors (VHTR). In: Encyclopedia of Nuclear Energy, vol. 1, pp. 507–517.

Hamon, D., 2021. Boiling Water Reactors. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 214–235.

Hatala, B., 2021. Gas Cooled Fast Reactors (GFR). In: Encyclopedia of Nuclear Energy, vol. 1, pp. 540–547.

Heidet, F., 2021. Sodium Cooled Fast Reactors. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 485–506.

Heuer, D., et al., 2014. Towards the thorium fuel cycle with molten salt fast reactors. Annals of Nuclear Energy 64, 421–429.

Hombourger, B., 2013. Parametric lattice study for conception of a molten salt reactor in closed thorium fuel cycle. Master's Thesis. Joint EPFL-ETHZ Master of Science in Nuclear Engineering.

Hombourger, B., 2018. Conceptual Design of a Sustainable Waste Burning Molten Salt Reactor. Doctoral Thesis. EPFL, Lausanne.

Hombourger, B., et al., 2015. Parametric lattice study of a graphite-moderated molten salt reactor. The ASME Journal of Nuclear Engineering and Radiation Science 1, 011009.
Hombourger, B., et al., 2016. The EQL0D procedure for fuel cycle studies in molten salt reactors. In: Proceedings of ICAPP 2016, San Francisco, USA.
Hombourger, B., Krepel, J., Pautz, A., 2019. Breed-and-burn fuel cycle in molten salt reactors, Progress in the Science and Technology of Nuclear Reactors using Molten Salts. EPJ Nuclear Sciences & Technologies 5, 10.
Huang, Y., Zang, J., 2021. Supercritical Water Cooled Reactors (SCWR). In: Encyclopedia of Nuclear Energy, vol. 1, pp. 564–576.
IAEA, 2013. Evaluation of High Temperature Gas Cooled Reactor Performance: Benchmark Analysis Related to the Pbmr-400, Pbmm, Gt-mhr, Htr-10 and the Astra Critical Facility. IAEA Tecdoc No. 1694. International Atomic Energy Agency.
IAEA, 2021. Status of Molten Salt Reactor Technology. In: IAEA Technical Report Series. International Atomic Energy Agency. Document In Preparation.
IGCAR, 2004. Prototype Fast Breeder Reactor Preliminary Safety Analysis Report. IGCAR.
Ignatiev, V., 2021. Molten Salt Reactors (MSR). In: Encyclopedia of Nuclear Energy, vol. 1, pp. 548–563.
Journet, J., et al., 1993. Minor actinides transmutation in oxide fuelled fast reactors. In: Proceeding of the GLOBAL93 Conference, Seattle, USA.
Krepel, J., 2021a. Self-Sustaining Breeding in Advanced Reactors: Characterization of Natural Resources. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 781–795.
Krepel, J., 2021b. Self-Sustaining Breeding in Advanced Reactors: Comparison of Fuel Cycle Performance. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 8151–831.
Krepel, J., Kramer, K.J., 2021. Molten Chloride Fast Reactors (MCFRs). In: Encyclopedia of Nuclear Energy, vol. 1, pp. 620–637.
Krepel, J., Losa, E., 2016. Enumeration of static and dynamics neutron consumption D-factor for several selected reactors at equilibrium closed fuel cycle. In: Proceedings of ICAPP 2016 Conference, San Francisco, USA.
Krepel, J., Losa, E., 2017. Comparison of fast reactors performance in the closed U-Pu and Th-U cycle. In: Proceedings of FR17, International Conference on Fast Reactors and Related Fuel Cycles, Yekaterinburg, Russian Federation, June 26–29.
Krepel, J., Losa, E., 2019. Closed U-Pu and Th-U cycle in sixteen selected reactors: Comparison of major equilibrium features. Annals of Nuclear Energy 128, 341–357.
Krepel, J., et al., 2008. Equilibrium cycle analysis of a gas cooled fast reactor with the EQL3D procedure. In: Proceedings of the International Conference PHYSOR 2008, Interlaken, Switzerland.
Krepel, J., et al., 2011. Comparison of safety related parameters of Generation-IV fast reactors in equilibrium closed cycle. In: Proceedings of the Global'11 Conference, Makuhari, Japan.
Krepel, J., Pelloni, S., Mikityuk, K., 2012. Comparison of open and closed U–Pu equilibrium fuel cycles for Generation-IV fast reactors with the EQL3D procedure. Nuclear Engineering and Design 250, 392–402.
Krepel, J., et al., 2014. Fuel cycle advantages and dynamics features of liquid fueled MSR. Annals of Nuclear Energy 64, 380–397.
Krepel, J., Hombourger, B., Losa, E., 2018. Fuel cycle sustainability of Molten Salt Reactor concepts in comparison with other selected reactors. In: Proceedings of the PHYTRA4 Conference, Marrakech, Morocco, September 17–19.
Leppänen, J., 2013. Serpent a continuous-energy Monte Carlo reactor physics burnup calculation code. In: VTT Technical Research Centre of Finland, Finland.
Losa, E., 2016. U-Pu and Th-U fuel cycle closure. PhD Thesis. Czech Technical University in Prague. Department of Nuclear Reactors, Prague.
MacPherson, H.G., 1985. The molten salt reactor adventure. Nuclear Science and Engineering 90, 374–380.
Mohapatra, D.K., et al., 2013. Physics aspects of metal fuelled fast reactors with thorium blanket. Nuclear Engineering and Design 265, 1232–1237.
Nichita, E., 2021. Heavy water reactors. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 236–251.
Nuttin, A., et al., 2005. Potential of thorium molten salt reactors detailed calculations and concept evolution with a view to large scale energy production. Progress in Nuclear Energy 46, 77–99.
OECD, 1995. LEU-COMP-THERM-060. In: International Criticality Safety Benchmark Evaluation Project (ICSBEP), Nuclear Energy Agency, NEA/NSC/DOC/(95)03/IV, Volume IV.
Perkó, Z., et al., 2015. Core neutronics characterization of the GFR2400 gas cooled fast reactor. Progress in Nuclear Energy 83, 460–481.
Fratoni, M., 2021. Fluoride Salt Cooled High Temperature Reactors. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 597–603.
Pounders, J.M., et al., 2011. A 3D stylized half-core CANDU benchmark problem. Annals of Nuclear Energy 38 (4), 876–896.
Rimpault, G., et al., 2002. The ERANOS code and data system for fast reactor neutronic analyses. In: Proceedings of the PHYSOR 2002 conference, Seoul, Korea, October 7–10.
Rosenthal, M.V., et al., 1971. Advances in the development of MSBR. In: Proceedings of the 4th International Conference on the Peaceful Uses of Atomic Energy, Geneva.
Salvatores, M., et al., 2004. The physics of TRU transmutation—A systematic approach to the intercomparison of systems. In: Proceedings of the PHYSOR 2004 Conference, Chicago, Illinois, April 25–29.
Schulenberg, T., 2012. High Performance Light Water Reactor: Design and Analyses. KIT Scientific Publishing.
Seifried, J.E., Gorman, P.M., Vujic, J.L., Greenspan, E., 2013. Accelerated equilibrium core composition search using a new MCNP-based simulator. In: Joint International Conference on Supercomputing in Nuclear Applications and Monte Carlo 2013 (SNA + MC 2013), Paris, France.
Shusterman, J., 2021. Fissile and Fertile Fuels. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 66–77.
Shwageraus, E., 2021. Light Water Cooled (LWR) Concepts. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 752–765.
Stanculescu, A., 2021. Worldwide Status of Advanced Reactors (GEN IV) Research and Technology Development. In: Encyclopedia of Nuclear Energy, vol. 1, pp. 473–484.
Taube, M., 1977. Fast reactors using molten chloride salts as fuel. In: INFCE Working Group 8, Advanced Fuel Cycle and Reactor Concepts, INFCE/DEP./WG.8/75.
Walter, A.E., Reynolds, A.B., 1982. Fast Breeder Reactors. Pergamon Press.